

The 2002 IAEA test spectra for low-level γ -ray spectrometry software

José M^a Los Arcos¹, Menno Blaauw², Stjepko Fazinic³, Vladimir P. Kolotov⁴

¹*Laboratorio de Patrones Nacionales, Asociado al C.E.M., CIEMAT, Avda. Complutense 22, Madrid 28040, Spain*

²*University of Technology Delft, Mekelweg 15, 2629 JB Delft, The Netherlands*

³*International Atomic Energy Agency, P.O. Box 100, Wagramerstrasse 5, A-1400 Wien, Austria, present address Rudjer Boskovic Institute, Bijenicka c. 54, 10000 Zagreb, Croatia*

⁴*Institute of Geochemistry and Analytical Chemistry of Russian Academy of Sciences, Kosygin str. 19, Moscow B-334, 117975 Russia*

Test spectra for low-level γ -ray spectrometry were acquired and made available to the general public at www.iri.tudelft.nl/~rc/fmr/iaea2002. As opposed to the 1995 test spectra, where reference values were made available only for the peak energies and areas, the new set of test spectra was acquired with certified sources, so that the reference values are radionuclide activities. Two well-defined detection geometries were employed: A 500 ml Marinelli beaker on a 33 % relative efficiency HPGe detector, and a 100 ml pillbox on a 96 % HPGe detector. The complete set addresses various issues that are especially important in low-level gamma-ray spectrometry, i.e. determination of efficiency curves in the presence of coincidence summing, differences in source geometry and density between standard and sample, poor statistics, shielding of background by the sample, use of low X- or γ -ray energies and the assumption of secular equilibrium of the natural radionuclides. The set was used in an IAEA intercomparison of software packages in December, 2002, reported on in a separate paper.

1. Introduction

In 1995, an intercomparison of gamma-ray spectrum analysis software based on a newly prepared set of test spectra was performed by the IAEA. The results, including the diskette with the test spectra, were published as an IAEA TECDOC-1011 "Intercomparison of gamma ray analysis software packages". In addition, two articles giving brief explanations of the test spectra, intercomparison method used and the results, were published in this journal [1,2].

The 1995 comparison focused on the ability of the programs to determine peak areas and energies with associated uncertainties. It was concluded that most programs yielded near-optimum results for singlet peaks, that all programs had difficulties with doublets, and that all programs yielded the same peak areas in spite of possible differences in peak shape models used.

However, good peak areas and energies are only the first step in obtaining measurement results in terms of radionuclide activities. If radionuclide-specific approaches are considered too laborious, efficiency curves are used.

It was therefore decided at the IAEA that a second set of γ -ray test spectra was required in order to assess the quality of an analysis program for low-level applications, this time consisting of matched sets of calibration and test spectra, with certified activities and detection conditions specified throughout.

In this paper, the design criteria and experimental procedures are described. The information given in the documentation supplied with the set of test spectra is more extensive.

2. Test set design considerations

The following issues were noted to be especially relevant in low-level gamma-ray spectrometry:

1. As a result of low activity levels and limited available counting times, environmental gamma-ray spectra often exhibit poor counting statistics. It is known from previous intercomparisons that some gamma-ray spectrometry programs have difficulty in detecting and properly quantifying the peak area and its uncertainty under this condition. This is also reflected in poor estimation of detection limits, false hits and misses.
2. Typically, since the samples tend to be voluminous and matrix density and composition is therefore relevant, the fact that calibration sources and samples may be different in this respect will be a source of possible error.

3. Samples will mostly be counted close to the detector, and the detector is likely to be highly efficient. As a result of this, the determination of the detector efficiency may be complicated by cascade summing effects. Also, the interpretation of a sample spectrum will be affected by the cascade summing. (These problems may be circumvented by radionuclide-specific calibration, but that is a laborious procedure.)
4. At low gamma-ray energies, say below 100 keV, detection efficiency strongly varies with photon energy. The methods used to construct the efficiency curve become very important here, especially when a curve is extrapolated.
5. In environmental measurements, the naturally occurring radionuclides are relevant. These may be present in the sample in secular equilibrium, but this is not necessarily the case, due to the sampling method or due to radiochemical separations. Analysis software should allow the user to gain insight in the equilibrium being disturbed or not.
6. Also in environmental measurements, the sample spectrum will show the same radionuclides as the background spectrum, and the count rates may be comparable. Background subtraction can therefore be a major source of error.
7. Voluminous samples will shield the detector from the surrounding materials, affecting the influence of the background.

Other considerations are:

- a) Sample geometries and detectors should be selected that are commonly used in routine practice.
- b) Different analysis programs to be tested will have special requirements as to the calibration spectra and other information needed. The test set should be designed in such a way that as many as possible of these special requirements are met.
- c) Not all programs will have provisions for cascade summing corrections. The test set should allow for such programs to correctly determine an efficiency curve and determine activities of radionuclides not exhibiting cascade summing effects.

3. Experimental

3.1. Sources

Four certified, NIST-traceable voluminous sources were purchased from Analytics. Inc., Oak Ridge, USA. Two were prepared in 0.1 l pillbox geometries, and two in 0.5 l Marinelli beakers. The sample container dimensions are shown in Figure 1 and Figure 2. The source compositions and activities are shown in Table 1, Table 2 and Table 3. All four sample containers were made of high-density polypropylene.

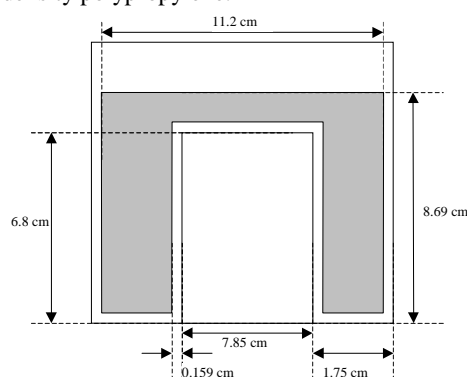


Figure 1: Marinelli beaker dimensions

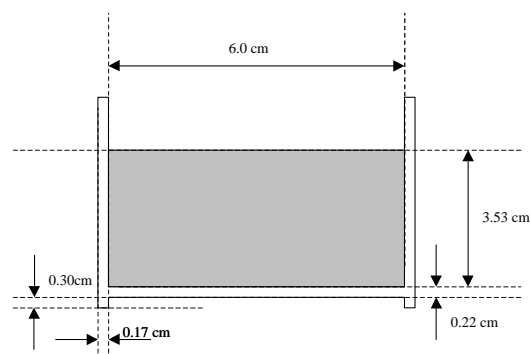


Figure 2: Pillbox geometry dimensions

Table 1 : Certified activities in “Analytics” (at Nov 1 2002, 12:00 EST) voluminous calibration sources denoted “MARIC” and “PIC”.

	Marinelli (1 g/cm ³)		PillBox (1 g/cm ³)	
	MARIC		PIC	
Nuclide	Activity (Bq)	Unc. (%)	Activity (Bq)	Unc. (%)
Am-241	3353	1.7	5403	1.7
Cd-109	39214	1.7	63190	1.7
Ce-139	1325	1.7	2136	1.7

Co-57	884	1.7	1424	1.7
Cs-134	5529	1.7	8909	1.7
Cs-137	1106	1.6	1782	1.6
Hg-203	2666	1.7	4297	1.7
Mn-54	1564	1.7	2519	1.7
Sn-113	2391	1.7	3853	1.7
Y-88	3914	1.7	6306	1.7
Zn-65	3191	1.7	5142	1.7

Table 2: Certified activities in the “Analytics” test sources denoted “MARIT” and “PIT”

	Marinelli (1g/cm ³)		Pillbox (1.6 g/cm ³)	
	MARIT		PIT	
Radionuclide	Activity (Bq)	Unc.(%)	Activity (Bq)	Unc. (%)
Ba-133	1713	1.7	1253	1.7
Co-60	1076	1.7	799	1.7
Eu-152	3739	1.7	2827	1.7
Cr-51	7903	1.7	7827	1.7
Na-22	1073	1.7	1063	1.7

Table 3: Elemental matrix composition (in mass %) of the "Analytics" sources

	MARIC, MARIT and PIC	PIT
ρ	1.15 g/cm ³	1.6 g/cm ³
C	72.1	40.5
H	6.0	2.9
O	21.9	35.6
Ca	-	21

The MARIC and PIC samples were intended as calibration samples, the MARIT and PIT to be treated as unknown samples.

Two mixtures of IAEA natural radioactivity reference materials in secular equilibrium, denoted “MIX1” and “MIX2”, were prepared at the IAEA’s laboratory in Seibersdorf. The certified activities are shown in Table 4. Upon receipt in Delft, the materials were placed in the same type of Marinelli beakers as used at Analytics Inc., but this time sealed with an inner lid of 3 mm thick Lucite glued in place with acrylic kit. One more of these beakers was filled with demineralized water.

Table 4: Certified activities in the IAEA MIX1 and MIX2 test sources. In view of the half lives involved, there is no reference data in the underlying IAEA certificates.

	Marinelli (1g/cm ³)		Marinelli (1g/cm ³)	
	MIX1		MIX2	
Radionuclide	Activity (Bq)	Unc.(%)	Activity (Bq)	Unc. (%)
K-40	2	25	1.16	25
Th-232	1024	1.4	601.3	1.3
U-235	43.4	0.5	72.5	0.4
U-238	928.3	0.3	1570.5	0.3

Furthermore a certified Amersham mixed calibration point source, denoted “QCD”, was used that contained Cd-109, Co-57, Ce-139, Hg-203, Sn-113, Sr-85, Cs-137, Y-88 and Co-60 as well as single-radionuclide sources containing Am-241, Cd-109, Cs-137, Zn-65, Sn-113, Mn-54 and Co-57. These point sources were needed to provide programs that might need such information with efficiencies measured in coincidence-free geometries and with peak-to-total ratios measured at a single photon energy.

3.2. Detectors

Two detectors were employed: A 33 % Canberra HPGe detector, denoted “SMALL”, and a 96.3 % Ortec HPGe detector, denoted “BIG”. A 10 cm thick, tight-fitting lead castle in all directions except the front shielded the BIG detector. The SMALL detector was not shielded at all.

The associated electronics as well as some specifications are shown in Table 5. The original datasheets and dimensional information obtained by other means are made available with the test spectra.

Table 5: Main detector and acquisition electronics specifications

	SMALL	BIG
relative efficiency	33 %	96.3 %
resolution at 1333 keV	1.84 keV	1.82 keV
HV	Ortec 459	Emetron EHV-6000
amplifier	Ortec 572	Ortec 571
ADC	Northern NS-623	Canberra Accuspec card

3.3. Measurements

Three kinds of spectra were acquired on each detector: Background spectra, calibration spectra and test spectra.

3.3.1. Background spectra

Since the presence of large samples shields the detector from the environment, two background spectra were acquired for the SMALL detector: One with nothing present near the detector, and one with a 500 ml Marinelli beaker filled with distilled water present. These two spectra are denoted BGSMALLPOINT and BGSMALLMARI. For the BIG detector, only one background spectrum was acquired, denoted BGBIGPOINT, because the expected count rates from the pill-box samples were much higher than from the background, and moreover originate from radionuclides not present in the background, as opposed to the natural radioactivity measurements in Marinelli beakers on the SMALL detector.

3.3.2. Calibration spectra

The six single-nuclide point sources were measured at 10 cm distance from the BIG and the SMALL detector and at 20 cm distance from the BIG detector.

The spectra were denoted Am241BIG and Am241SMALL for the Am-241 source, likewise for the others.

The Amersham QCD point source was counted at 30 cm and 20 cm from the BIG detector, as well as on the end cap, and at 20 cm and 10 cm from the SMALL detector, as well as on the end cap. The spectra are denoted QCDBIG30, QCDBIG20, QCDBIG0, QCDSMALL20, QCDSMALL10 and QCDSMALL0.

The PIC source was counted on the BIG detector until 1 % precision was obtained in the net area of the Cs-134 sum peak at 1174 keV. The spectrum is denoted PICBIG.

The MARIC source was counted on the SMALL detector until 1 % precision was obtained in the net area of the Cs-134 sum peak at 1174 keV. The spectrum is denoted MARICSMALL.

3.3.3. Test spectra

The MIX1 and MIX2 samples were counted immediately after sealing as well as after three weeks decay time on the SMALL detector. Right after sealing, MIX1 was counted during 2 days. MIX2 was given a counting time of 23h35 to get excellent statistics, as well as short acquisition times of 20 and 6 minutes to get poor statistics. The short acquisition times were divided in two halves, one half counted just before the long count and one immediately after, so changes in mother-daughter equilibria should have a minor effect on the expected peak area ratios between excellent and poor statistics spectra.

After three weeks, more measurements were done for MIX1 and MIX2. Both were counted during 1 hour on the SMALL detector, and MIX2 was counted for an additional 3 minutes to get a “poor statistics” spectrum.

The spectra are denoted MIX1NEQLONG, MIX2NEQLONG, MIX2NEQ20, MIX2NEQ6, all before the Rn-222 equilibrium was achieved, and MIX1EQ, MIX2EQ and MIX2EQ3, after the equilibrium was achieved.

At Analytics Inc., a Marinelli beaker of the same dimensions and matrix density as the calibration one was prepared, and a 100 ml pillbox sample of higher density (1.6 g/ml) than the calibration one. The pillbox sample was counted on the BIG detector for the same duration as the pillbox calibration source, and also during 1 hour. The spectra are denoted PITBIG and PITBIG60. The Marinelli beaker was counted on the SMALL detector for the same duration as the calibration source, and also during 1 hour. The spectra are denoted MARITSMALL and MARITSMALL60.

4. Data handling

The MIX2NEQLONG spectrum was analysed with the in-house IRI software, knowing and using the radionuclides and peak energies involved, to get reference peak areas and energies. The same spectrum was also analyzed with the Interwinner program, again using all available information. The resulting two lists of peak areas were compared side by side. In those cases where the two peak areas agreed to within three times the uncertainty in the difference, i.e. $z < 3$, the unweighted averages of the two peak energies, the two areas and their uncertainties were adopted as the reference values. The z -score was calculated with

$$z = \frac{A_{\text{IRI}} - A_{\text{Interwinner}}}{\sqrt{e_{\text{IRI}}^2 + e_{\text{Interwinner}}^2}}$$

where A denotes peak area, and e its 1 s.d. uncertainty.

Cases where $z > 3$ were inspected visually and discussed. For the peaks at 46.4, 63.2, 77.1, 79.4, 81.1, 329.8, and 1764.5 keV, the Interwinner results were adopted. For the peaks at 405.1, 609.2, 768.2 and 1460.6 keV, the IRI results were adopted. The 238.6 keV was treated as if $z < 3$, even though the actual z -score was 6.2. The peaks at 609.2, 768.2, and 839.3 keV had been fitted as doublets by Interwinner but as singlets by the IRI software. After adding the Interwinner peak areas, they agreed with the IRI result and the averages were once again adopted as reference peak areas. Finally, all peak areas were divided by the ratio of counting times of the MIX2NEQLONG over MIX2NEQ6 spectra, i.e. 236.59, to obtain reference expected peak areas for the MIX2NEQ6 spectrum.

5. Relations between design criteria and actual spectra

The MIX2NEQ6 spectrum addresses issue 1 (poor statistics). The expected, reference areas are known from the MIX2NEQLONG spectrum. The latter can be analyzed and corrected for background with the BGSMAALLMARI spectrum, and the results interpreted in terms of radionuclide activities to verify proper background correction (issue 6 and 7).

The MARICSMALL and PICBIG spectra contain radionuclides that exhibit no coincidence summing, radionuclides that exhibit some coincidence summing (Co-60 and Y-88), and a radionuclide that exhibits extreme coincidence summing (Cs-134). By simply ignoring the latter radionuclides, a correct efficiency curve up 1115 keV can be obtained even by the simplest of programs. The most advanced ones can try their luck using the complete list of certified activities (issue 3). Once a correct efficiency curve has been obtained, any program should be able to come up with the correct activities in the MARITSMALL measurement for the simple radionuclides, but getting the right answer for Eu-152 and Co-60 requires coincidence-summing corrections, taking the voluminous effect into account.

The PITBIG spectrum can only be interpreted correctly after some correction has been applied to the efficiency curve for the higher density of the sample (issue 2). Once that has been done, correct results can be obtained for the coincidence-free radionuclides. The quality of the results obtained for the remaining radionuclides will depend on the abilities of the analysis program with respect to coincidence summing and the voluminous true-coincidence summing effect.

The “QCD” point-source calibration spectrum is provided for those programs that need to translate efficiency curves measured in a so-called “reference” geometry to the actual sample geometry. The other, single-radionuclide point source spectra allow for the determination of a peak-to-total ratio efficiency curve as is required for coincidence corrections by most of the available programs.

6. Discussion and conclusions

We believe - and the accompanying paper on intercomparison results demonstrates - that the set of test spectra presented in this paper allows for testing of a wide range of γ -ray spectrum analysis software, from programs that merely offer the determination of a full-energy peak efficiency curve for true-coincidence-free geometries up to programs that allow one to convert point-source efficiencies to efficiencies for samples of arbitrary composition and dimensions, including programs that take the voluminous true-coincidence summing effect into account. Any program, that can determine all the correct radionuclide activities in the test samples from the calibration sample spectra and the associated data, can be applied in practice with confidence.

The only issue we are aware of not specifically having addressed is the effect of angular correlations on true coincidence summing. These effects will be smaller than 5 % in practice. Also, since the spectra were obtained from real sources, these effects are present in the spectra and software developers and testers can specifically focus on those when using the set.

The set of test spectra with reference peak areas, extensive information on the certified activities as well as on the detector geometries is available via www.iri.tudelft.nl/~blaauw.

7. References

1. M. Blaauw, V. Osorio Fernandez, W. Westmeier, Nucl.Instr.Meth. A387 (1997) 410
2. M. Blaauw, V. Osorio Fernandez, P. van Espen, G. Bernasconi, R. Capote Noy, H. Manh Dung, N.I. Molla, Nucl.Instr.Meth. A387 (1997) 416